R-FLICS Meeting

Self-Assembled Materials Systems and Devices for R-FLICS

Prof. Seng-Tiong Ho, ECE Dept

Prof. Tobin Marks, Dept of Chemistry

Northwestern University

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OUTLINE

- Introduction
- Self Assembled Superlattice Material
- EO Device Fabrication
- Future Work

Motivation:

Next-Generation EO modulators

- $LiNO_3$ EO modulators with 10Gbit/sec data transfer rate are being used in current optical communication systems.
- Communication industries have identified 20-40 Gbit/sec as the requirement for next-generation EO modulators.
- The driving voltage should below 5 volt range at 20-40 GHz.
- Current EO modulators are based on bulk-grown $LiNO_3$ and have reached close to their performance limits.
- Novel approaches are required to realize the next-generation of EO modulators with modulation bandwidths of 20-40 GHz and above.

Approaches

Self-Assembled Supperlattice Chormophore EO Modulator for low Voltage and high frequency applications

POTENTIAL ADVANTAGES OF ORGANIC MODULATORS

A. **High frequency modulation can be achieved because of low ε.**

B. **Organic chromophores may be engineered to have High EO coefficients, leading to low switching voltage.**

C. **May be lower in cost**

SELF-ASSEMBLED CHROMOPHORES -- WHY POLED POLYMERS HAVE PROBLMES?

1. Poling does not give a large degree of polar alignment.

2. Poling:
   a. causes electrical damage to polymers;
   b. causes optical loss (hetergeneities & charge injection);
   c. poled systems are thermodynamically unstable (want to relax back);
   d. difficult manufacturing (difficult to pole uniformly reproducibly);
   e. electric field between poling electrodes can mechnically distort the waveguide.

SELF-ASSEMBLED SUPERLATTICE CHROMOPHORE

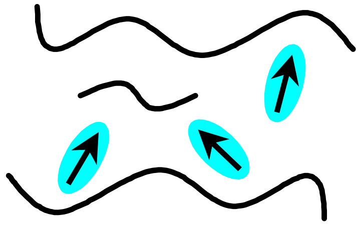
A. No poling required.

B. Can achieve high polar alignment, giving very large EO coefficient (i.e. low operation voltage).

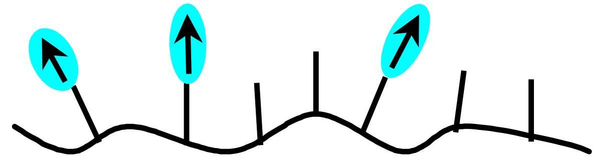
C. High frequency modulation can be achieved because of low ε.

DESIGN MOTIFS FOR MOLECULAR/POLYMER ELECTRO-OPTIC MATERIALS

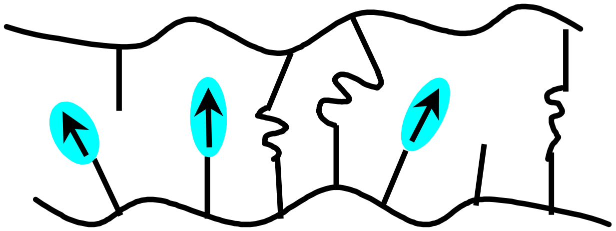
Poled Host-Guest



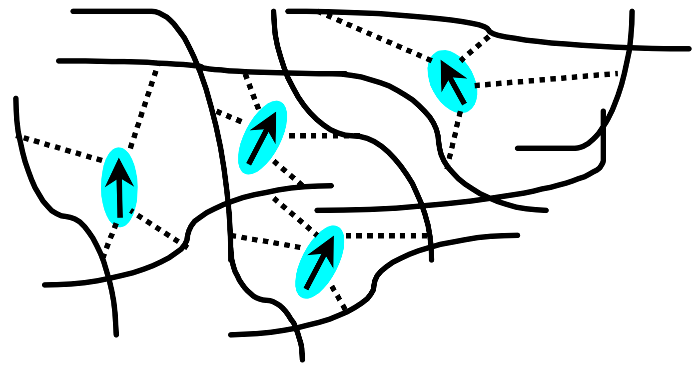
Poled and Functionalized



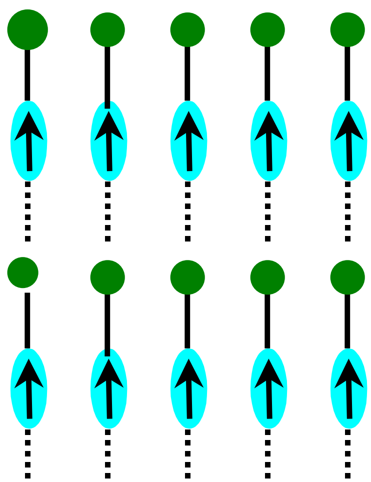
Poled, Functionalized and Crosslinked



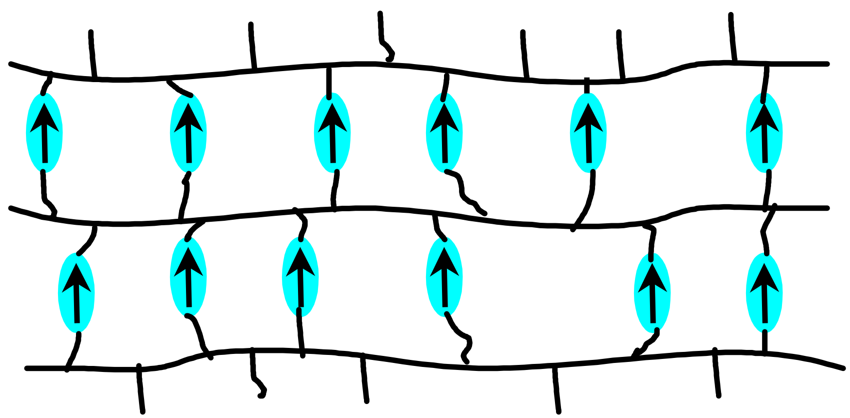
Poled, Crosslinkable Matrix



Chromophoric LB Film



Self-Assembled Superlattice (SAS)



 = Chromophore Module

Electro-Optic Modulators

Figures of Merit

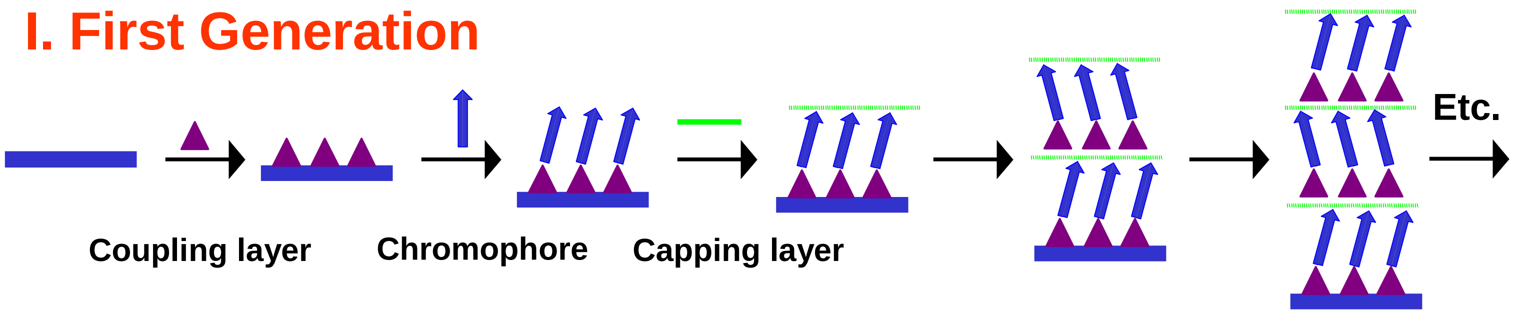
	$LiNbO_3$	EO Polymers	SA Materials
EO coefficient r (pm/V)	31	10-75	30-200
Dielectric constant ε	28	4	~6
Refractive Index n	2.2	1.6	1.6
n^3r (pm/V)	248	150	120-820
n^3r/ε	8.7	38	20-140

MOLECULAR SELF-ASSEMBLY OF HIGH PERFORMANCE ELECTRO-OPTIC STRUCTURES

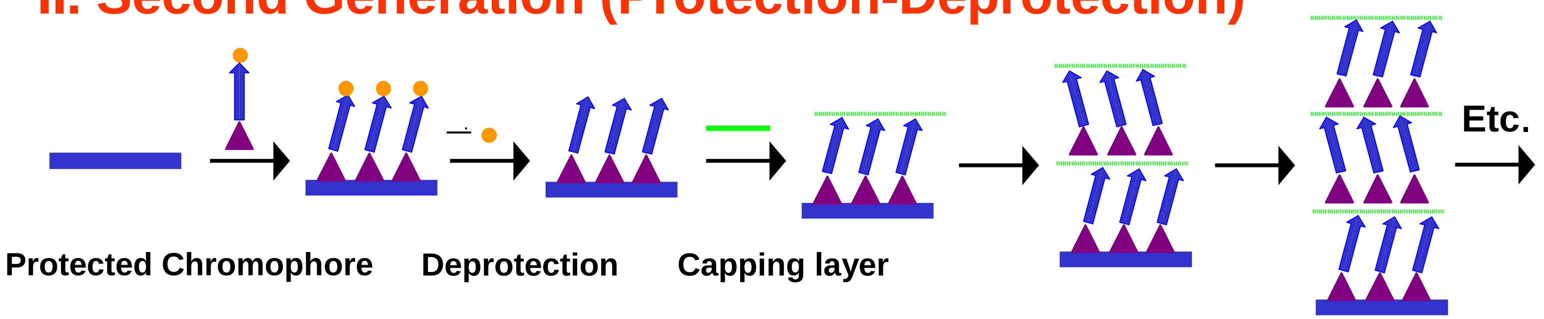
- Programmed Polar Microstructure
- Tailored Building Blocks
- Compatible with Soft Lithography
- n^3r/ε = 20-140 pm/V

- Synthetic Scope, Fidelity, Scalability
- Tune λ, β, r
- Templated Growth, Device Integration
- Microstructure, Loss

I. First Generation



II. Second Generation (Protection-Deprotection)

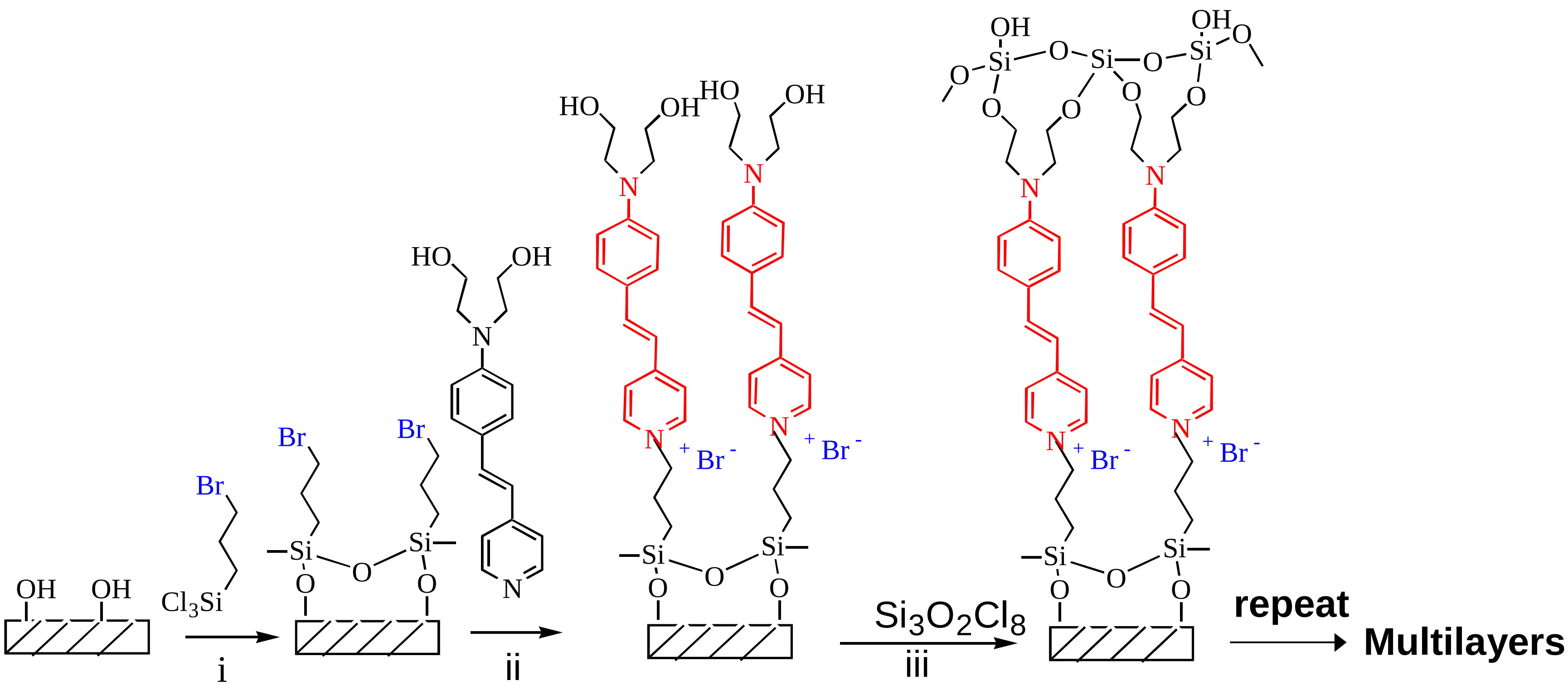


Rapid. Readily Adaptable to Automation

Robust, Adherent, Smooth, Structurally, Regular Siloxane Networks

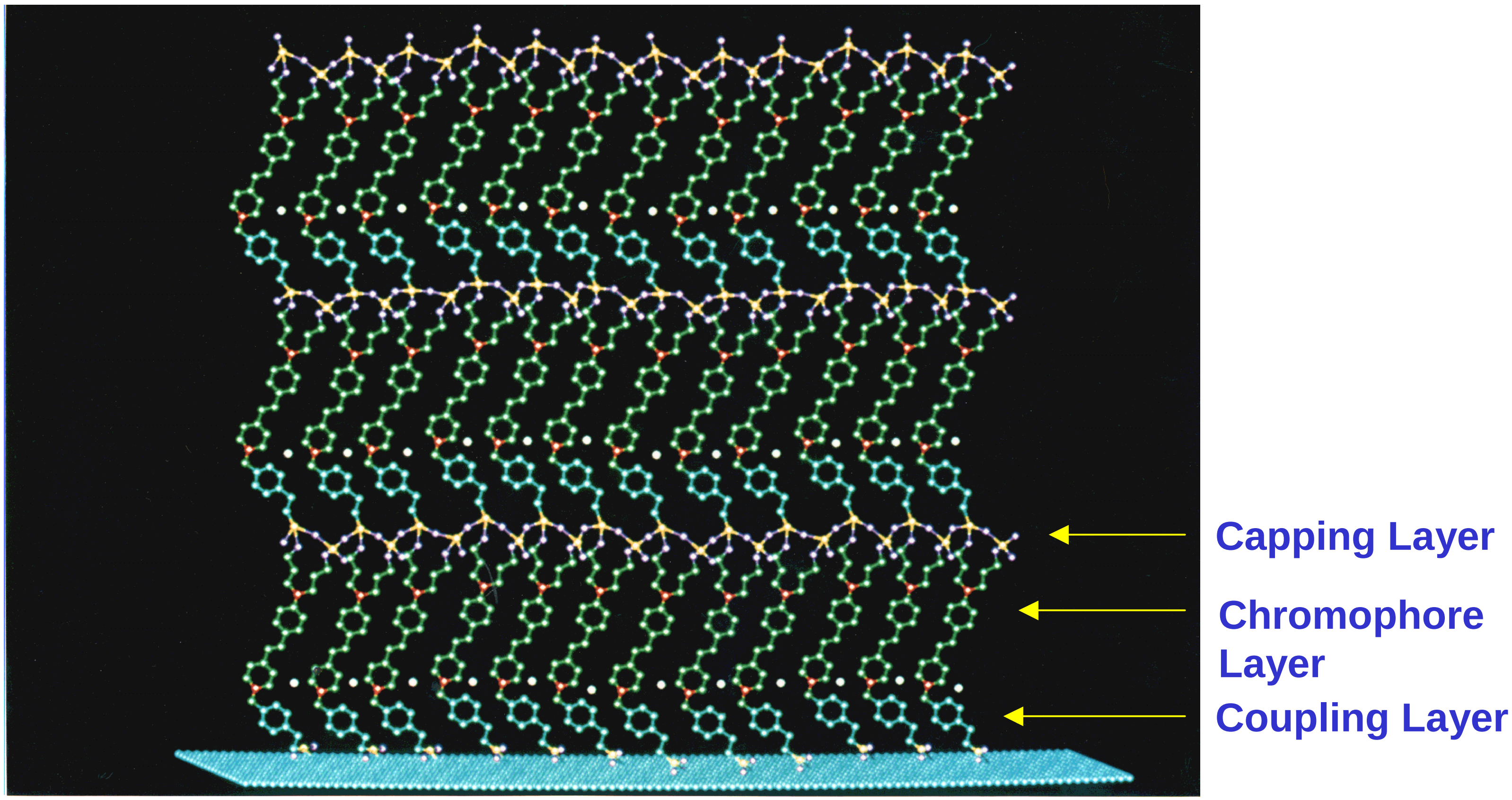
Construction of Chromophoric Multilayers by Molecular Layer Epitaxy

First Generation Self-Assembly

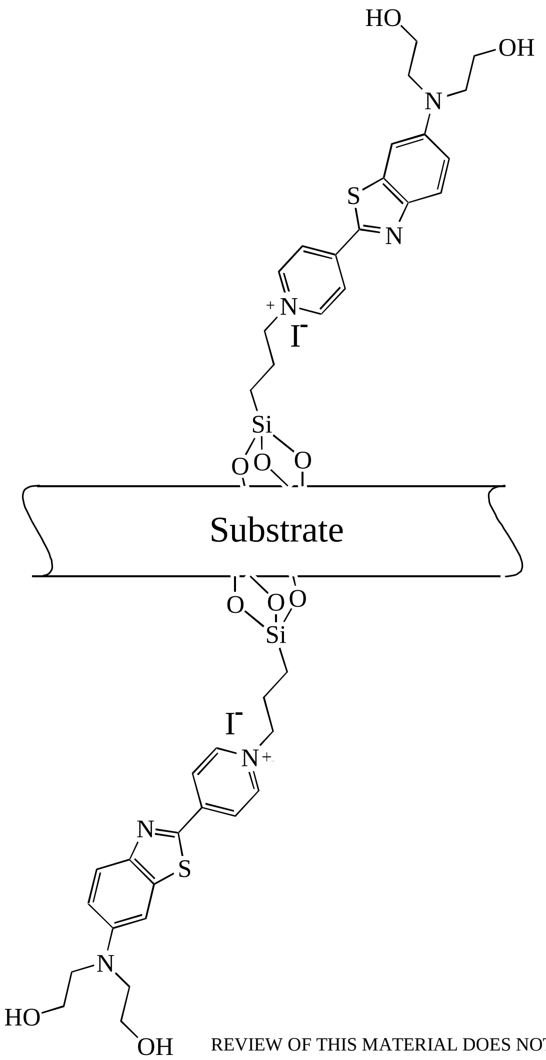


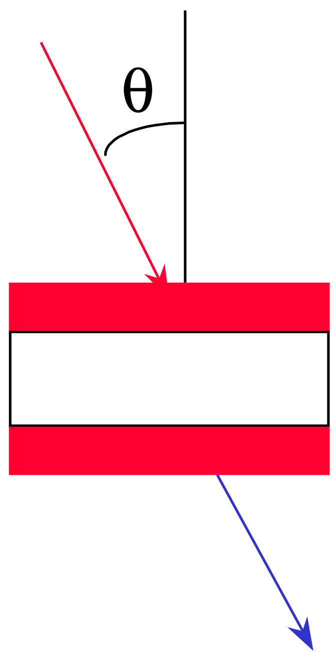
1. Rapid topotactic multilayer growth
2. Intrinsically acentric (no poling required)
3. Very high structural regularity
4. Very large $\chi^{(2)}$ response

MOLECULAR MODEL OF A CHROMOPHORIC SUPERLATTICE



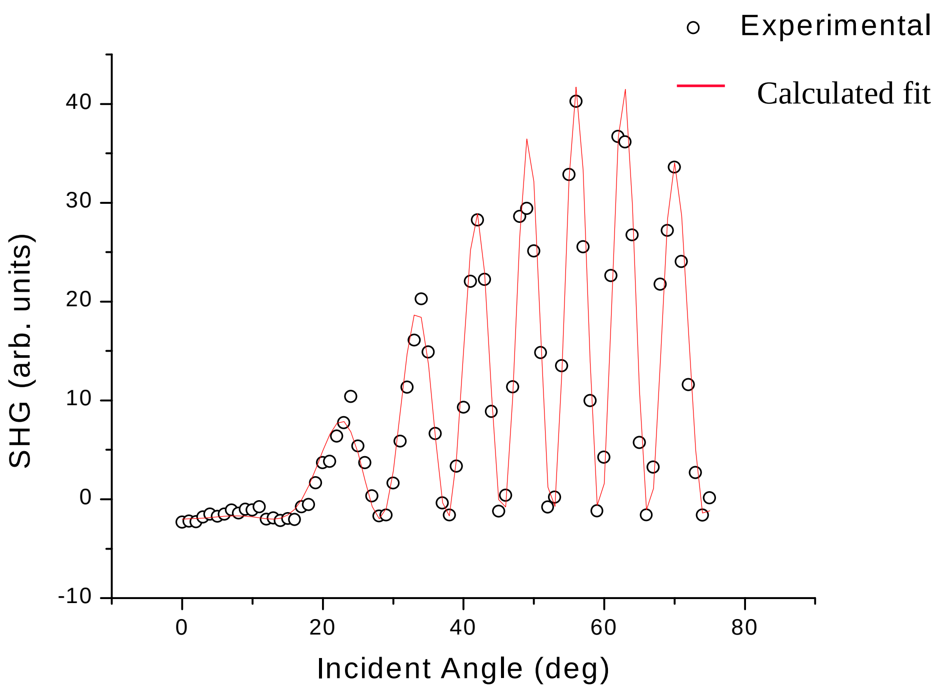
Determination of NLO Response for Self-Assembled Benzothiazole-Type Chromphores



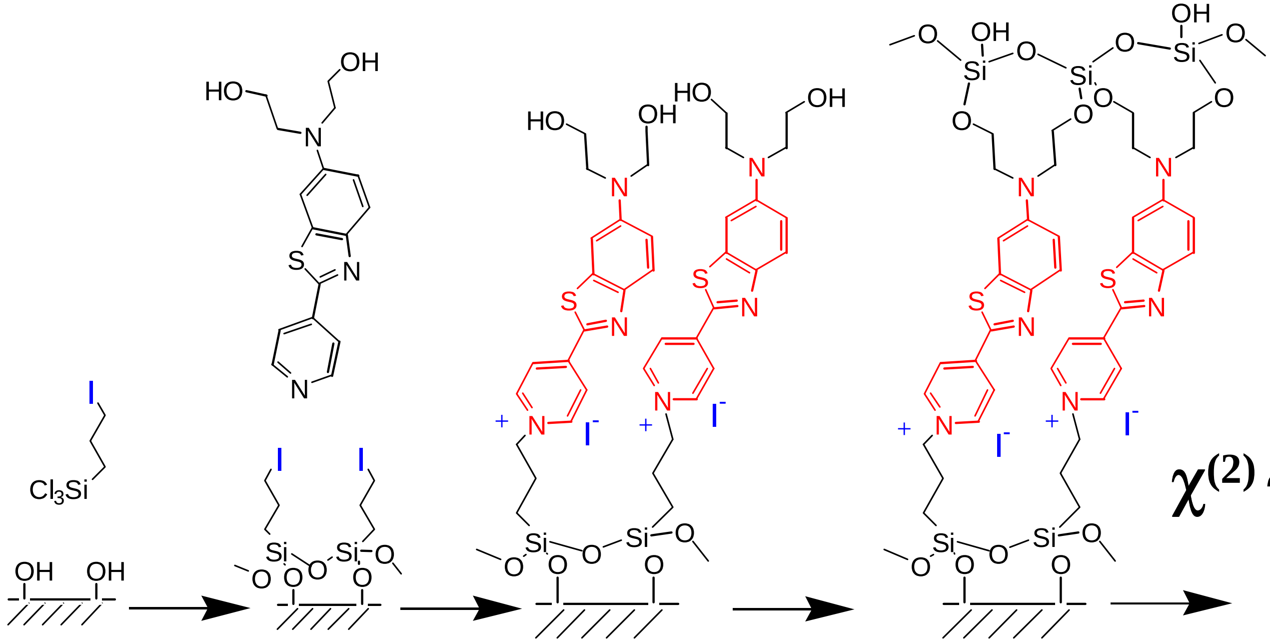


$$\chi^{(2)}_{zzz} = 334 \text{ pm/V @ 1064 nm}$$

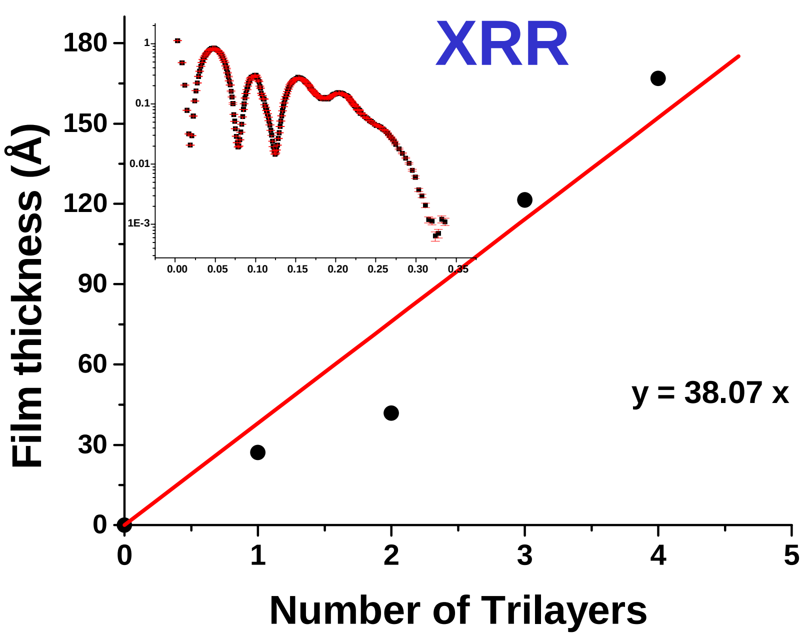
Tilt angle = 45°

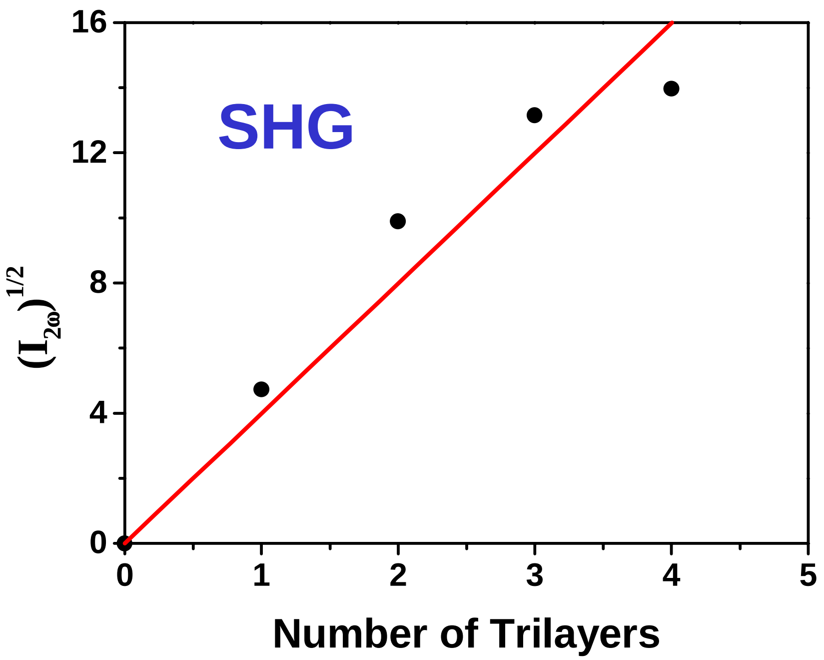


First Generation Self-Assembly



$\chi^{(2)} \sim 334$ pm/V





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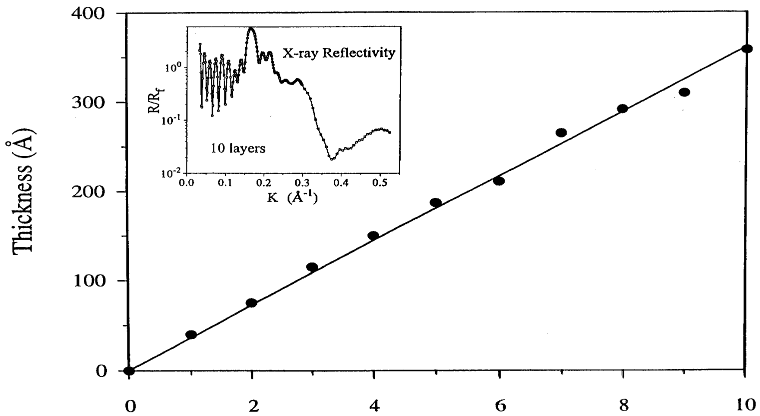
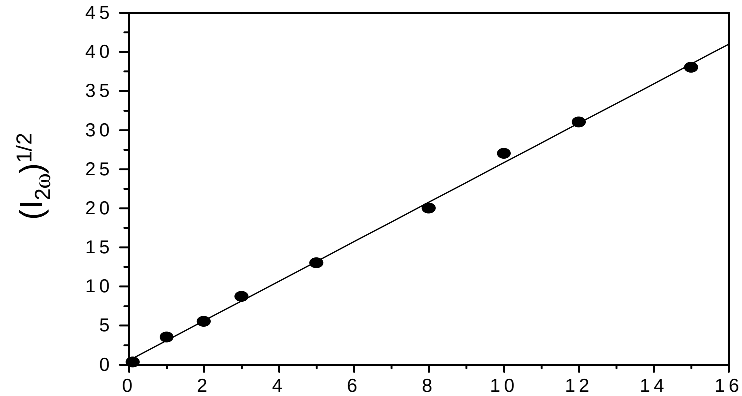
SELF-ASSEMBLED ELECTRO-OPTIC MATERIALS PROPERTIES

Microstructural Regularity (XRR)



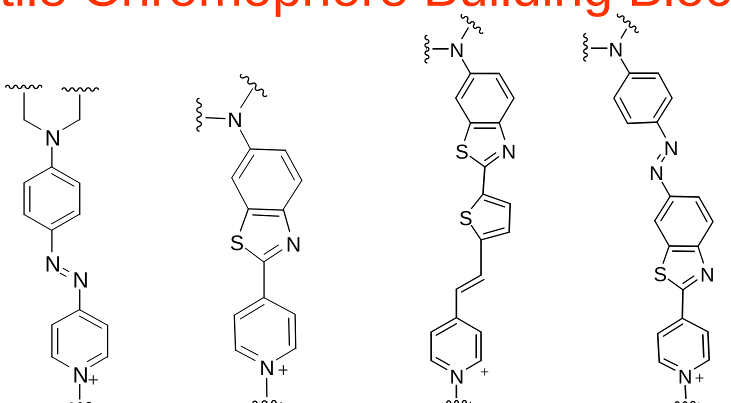
Number of Trilayers

Polar Regularity (SHG)



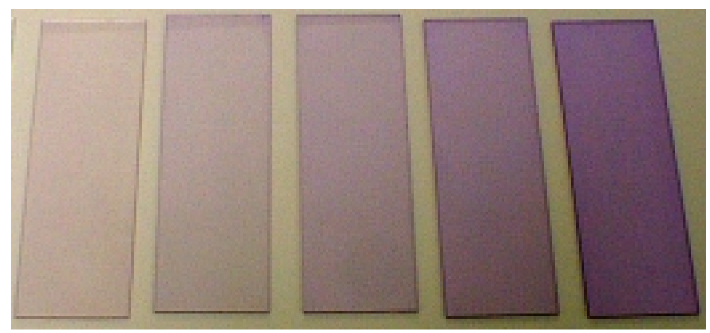
Number of Trilayers

Versatile Chromophore Building Blocks



β (0.65 eV)calcd. ($10^{-30}cm^5esu^{-1}$)	178	360	1288	1617
λ_{max}, calcd. (nm)	572	498	658	666
Film r_{33}, ω_o=1064 nm (pm/V)	56	125	410 (est.)	525 (est.)

Samples of Self-Assembled Films



1 2 3 4 5

Number of Trilayers

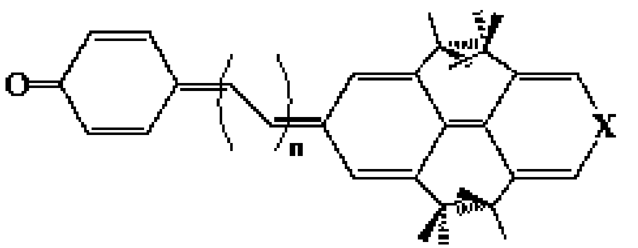
Ho ***Dutta*** ***Marks*** ***Ratner***

ELECTRONIC STRUCTURE THEORY IN MATERIALS DEVELOPMENT

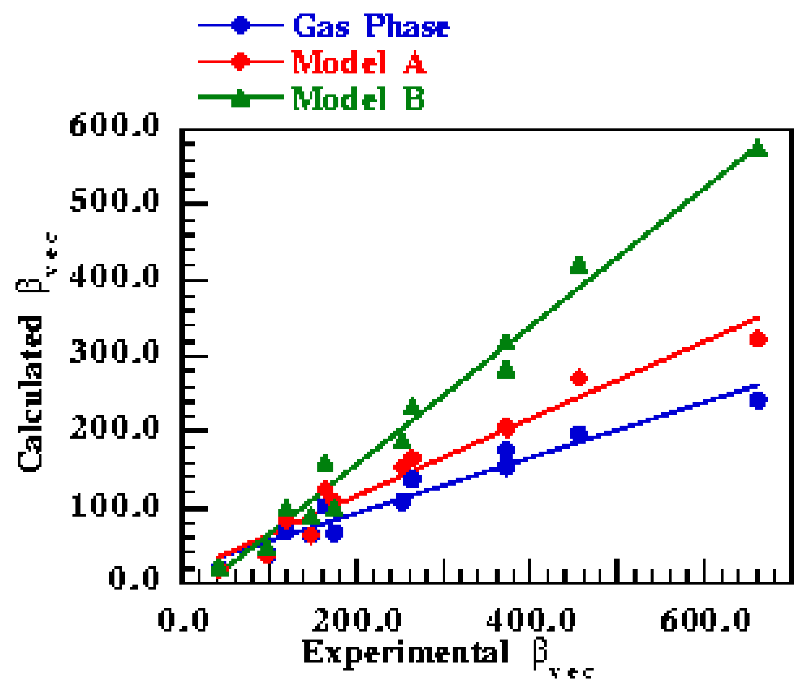
Correction Vector/Sum-Over-States ZINDO Calculations

ATTRACTIONS

- Target New Molecular Architectures For Synthesis
- Test New Response Mechanisms
- Understand Mechanisms, Frequency Dependence



$\mu\beta(0.65\ \mathrm{eV}) = 200{,}000 \times 10^{-48}$ esu



CHALLENGES

- Environmental Effects
- Metal-Organic Structures
- Open Shell Molecules, Excites States
- Luminescent Electron-Hole Recombination

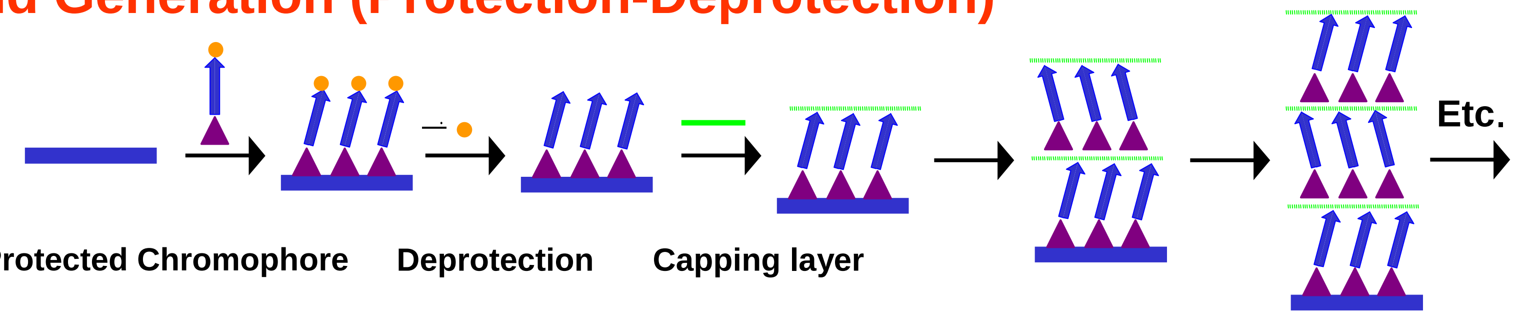
M. Ratner
I. Fragala
S. Di Bella

MOLECULAR SELF-ASSEMBLY OF HIGH REFRACTION INDEX ELECTRO-OPTIC STRUCTURES

- Programmed Polar Microstructure
- Tailored Building Blocks
- Compatible with Soft Lithography
- n^3r/ε = 20-140 pm/V

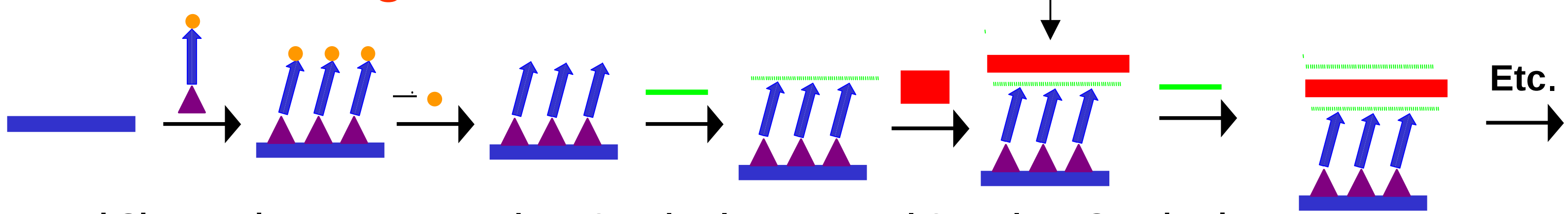
- Synthetic Scope, Fidelity, Scalability
- Tune λ, β, r
- Templated Growth, Device Integration
- Microstructure, Loss

Second Generation (Protection-Deprotection)



Metal Oxide Layer

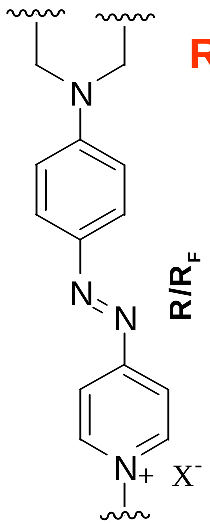
SA-Films with High Refraction Index



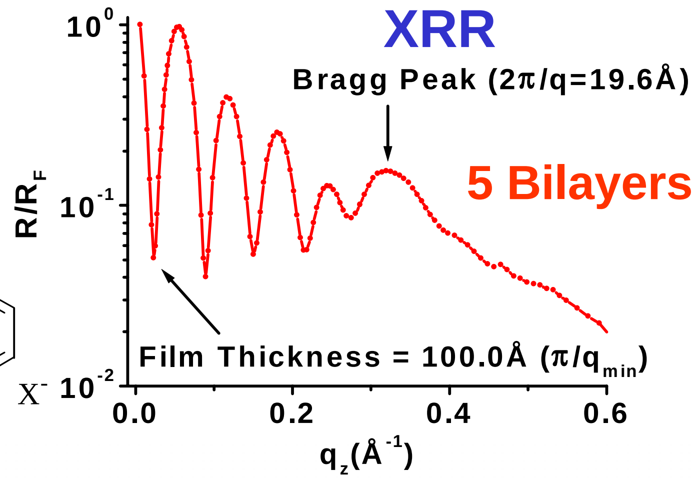
For First and Second Generation Self-Assembly

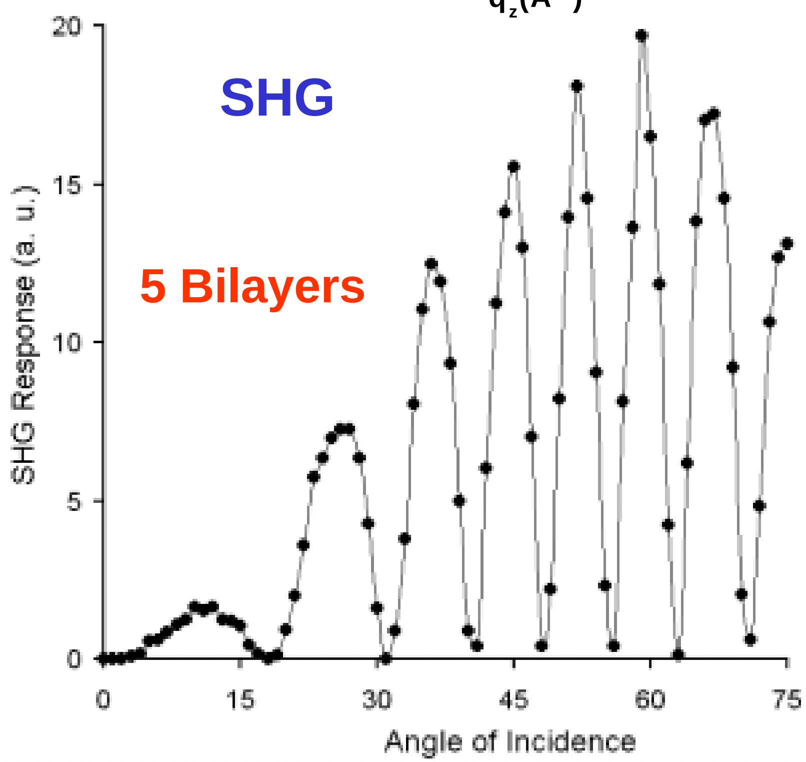
‘BIFUNCTIONAL HYBRID STRUCTURE’

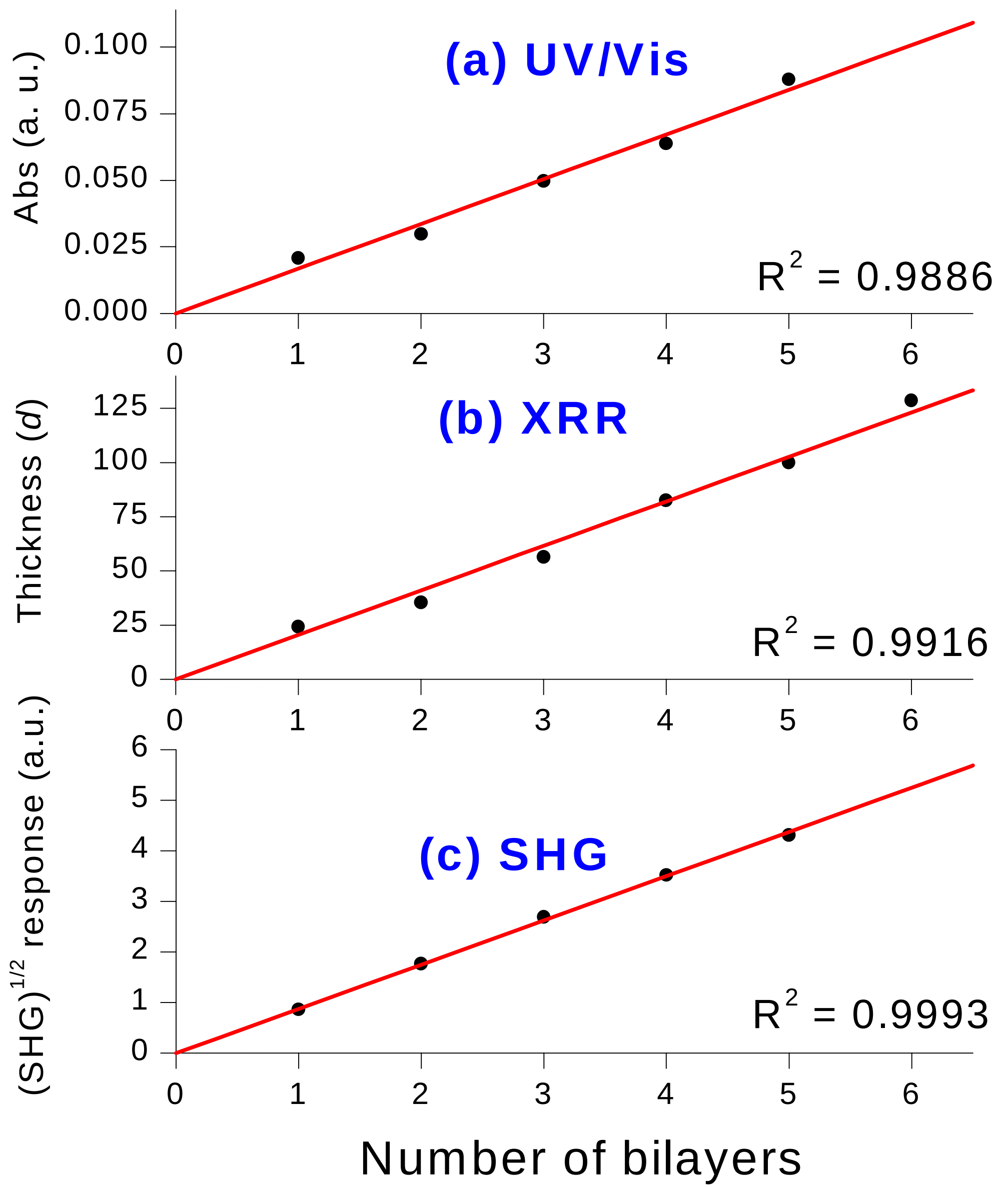
Second Generation Self-Assembly (Protection-Deprotection)



Readily Adaptable to Automation

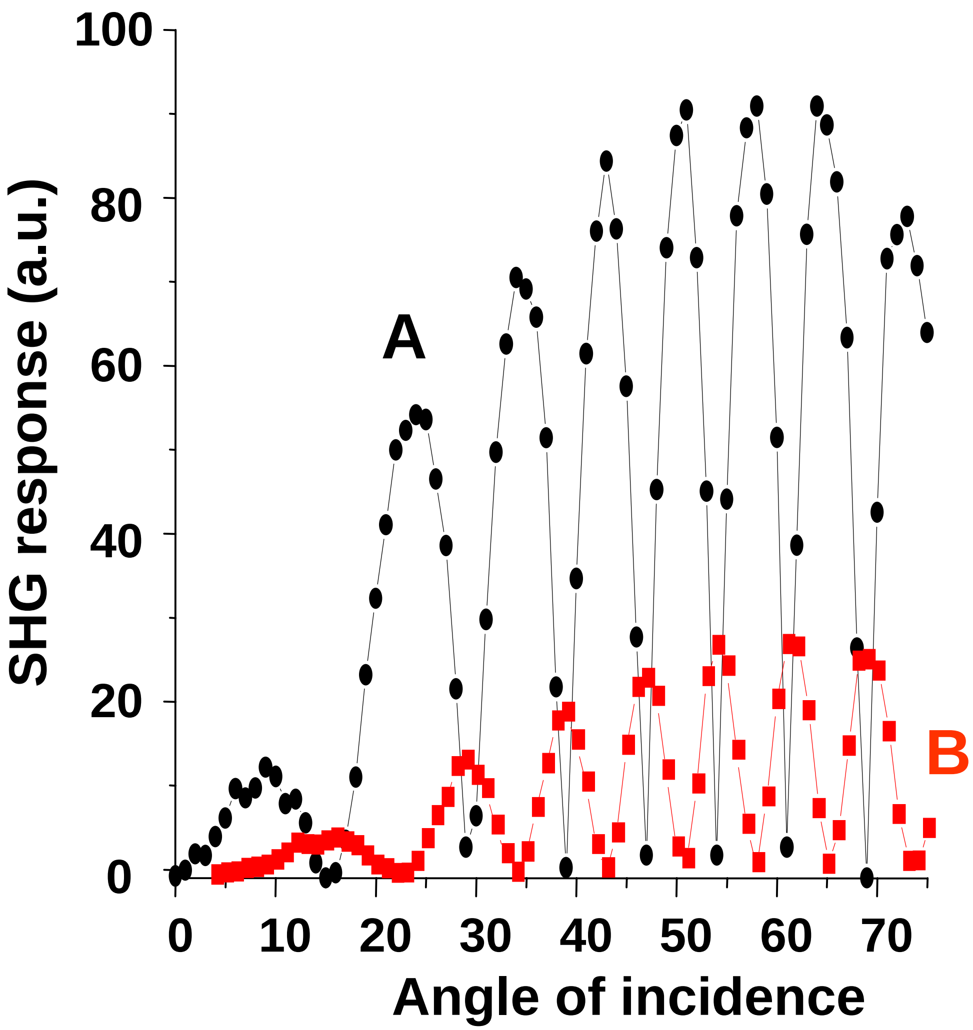


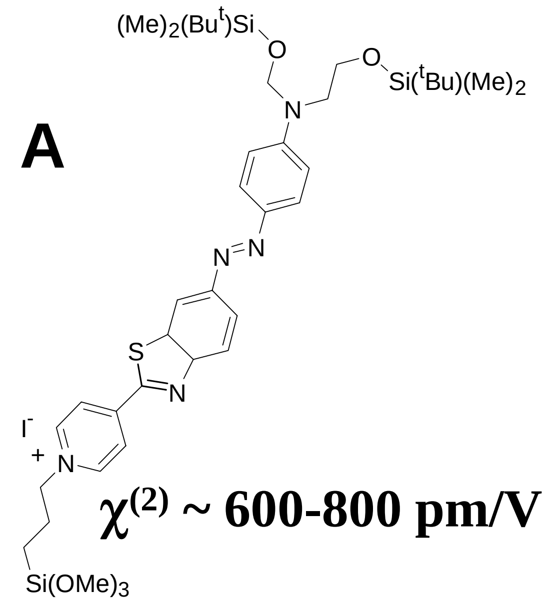




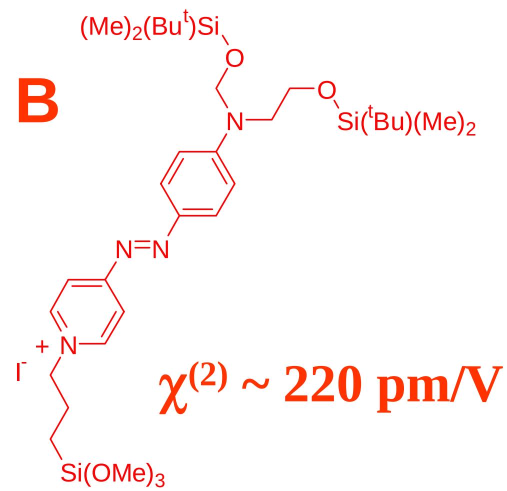
Comparison of NLO properties of thin films

Second Generation Self-Assembly





$\chi^{(2)} \sim 600\text{-}800$ pm/V



$\chi^{(2)} \sim 220$ pm/V

Second Generation Self-Assembly (Protection-Deprotection)

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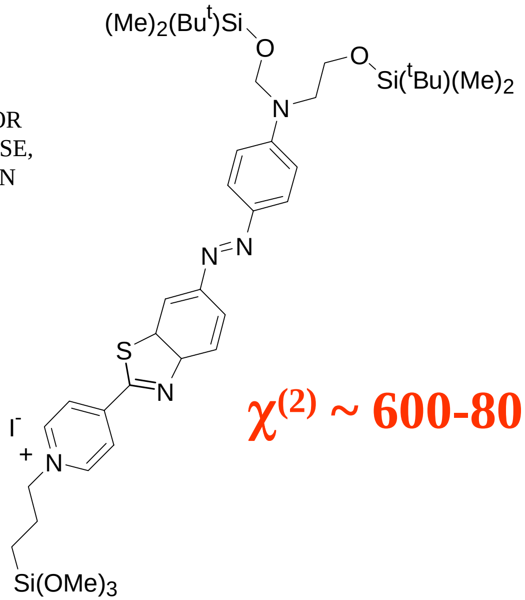
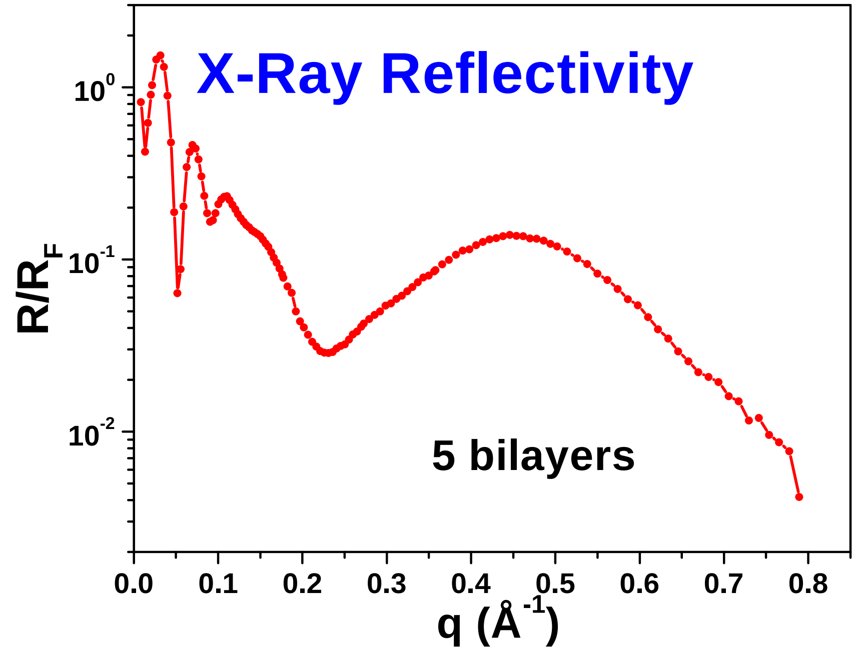
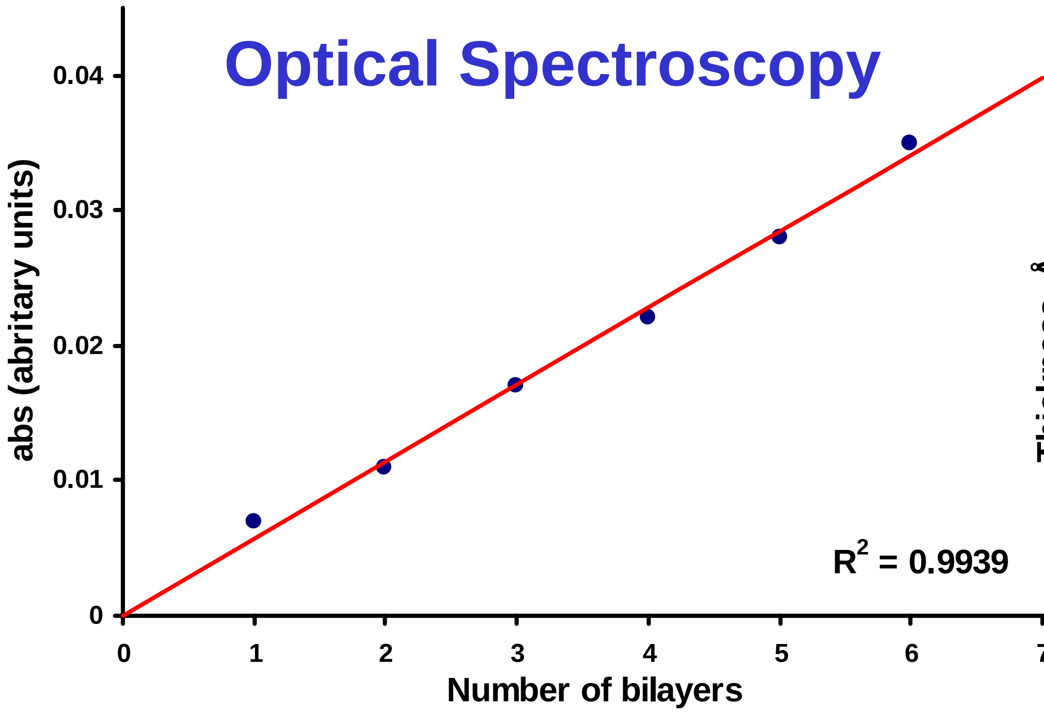
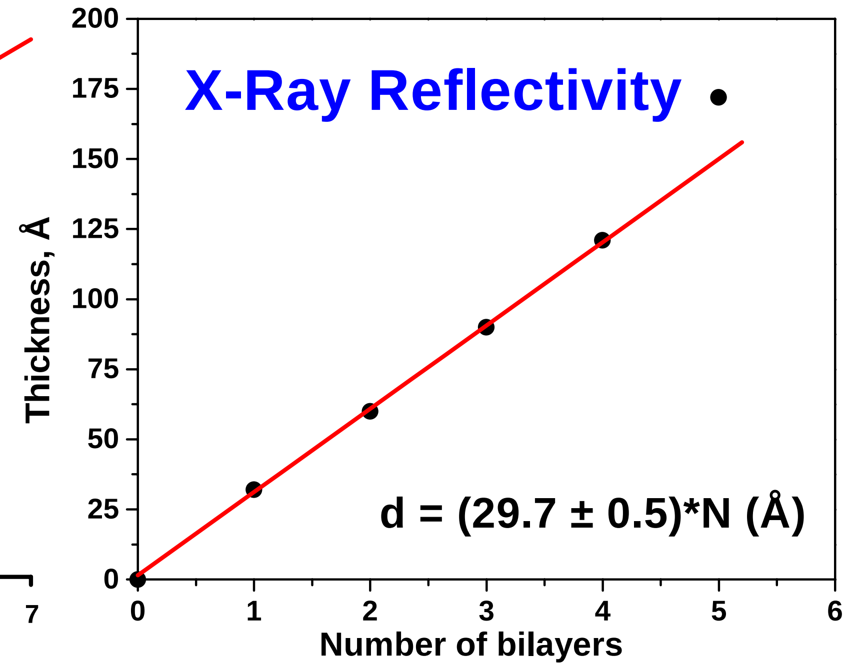


$\chi^{(2)} \sim$ 600-800 pm/V

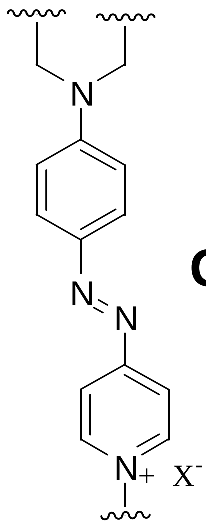


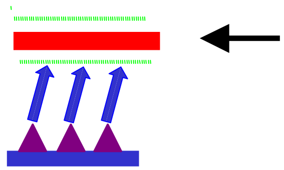




Second Generation Self-Assembly (Protection-Deprotection)

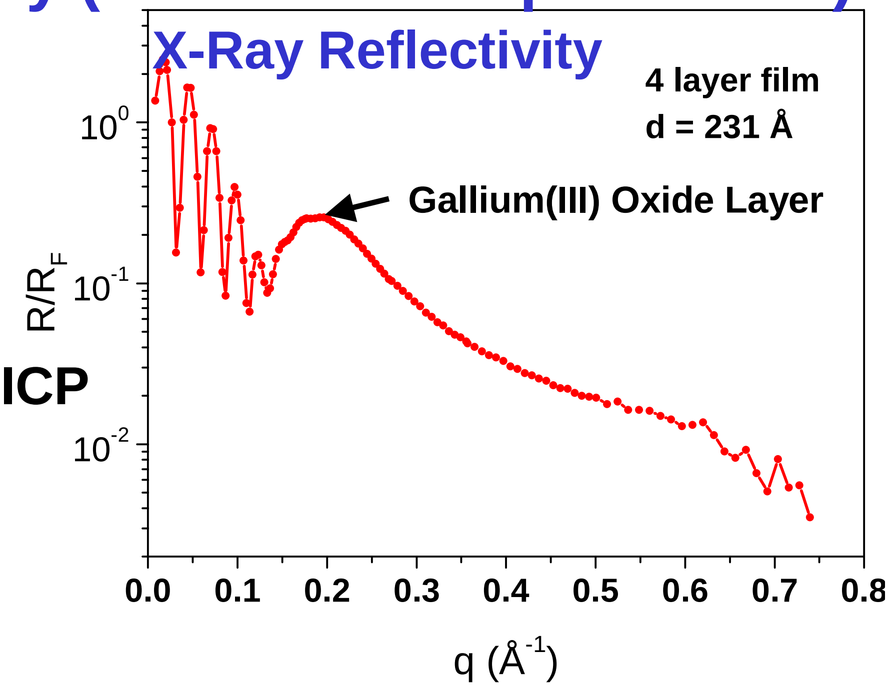
SA-Films with High Refraction Index



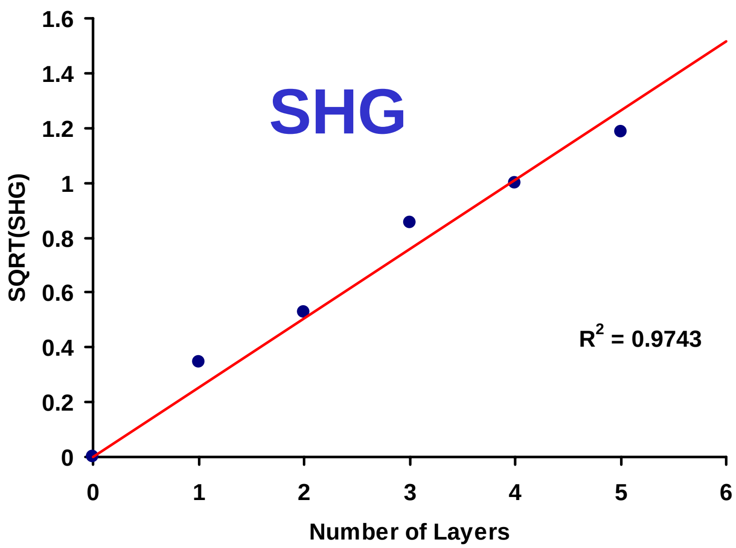


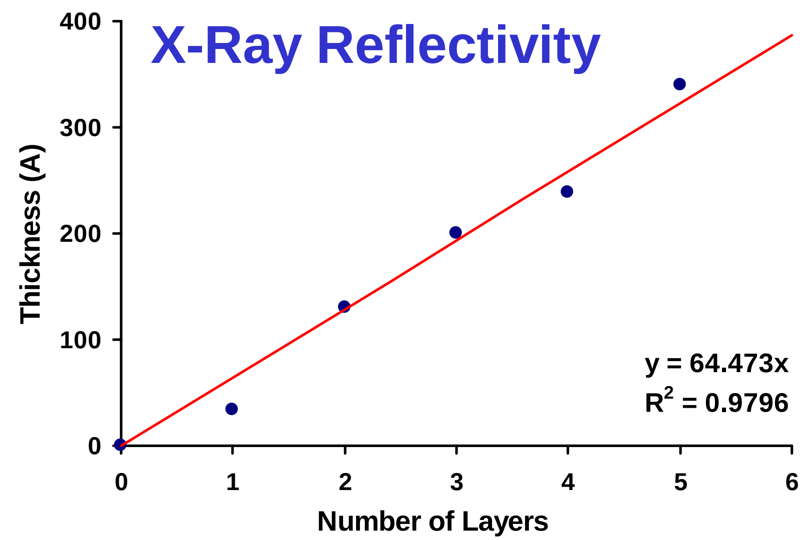
Gallium Oxide

Gallium observed by XRR and ICP



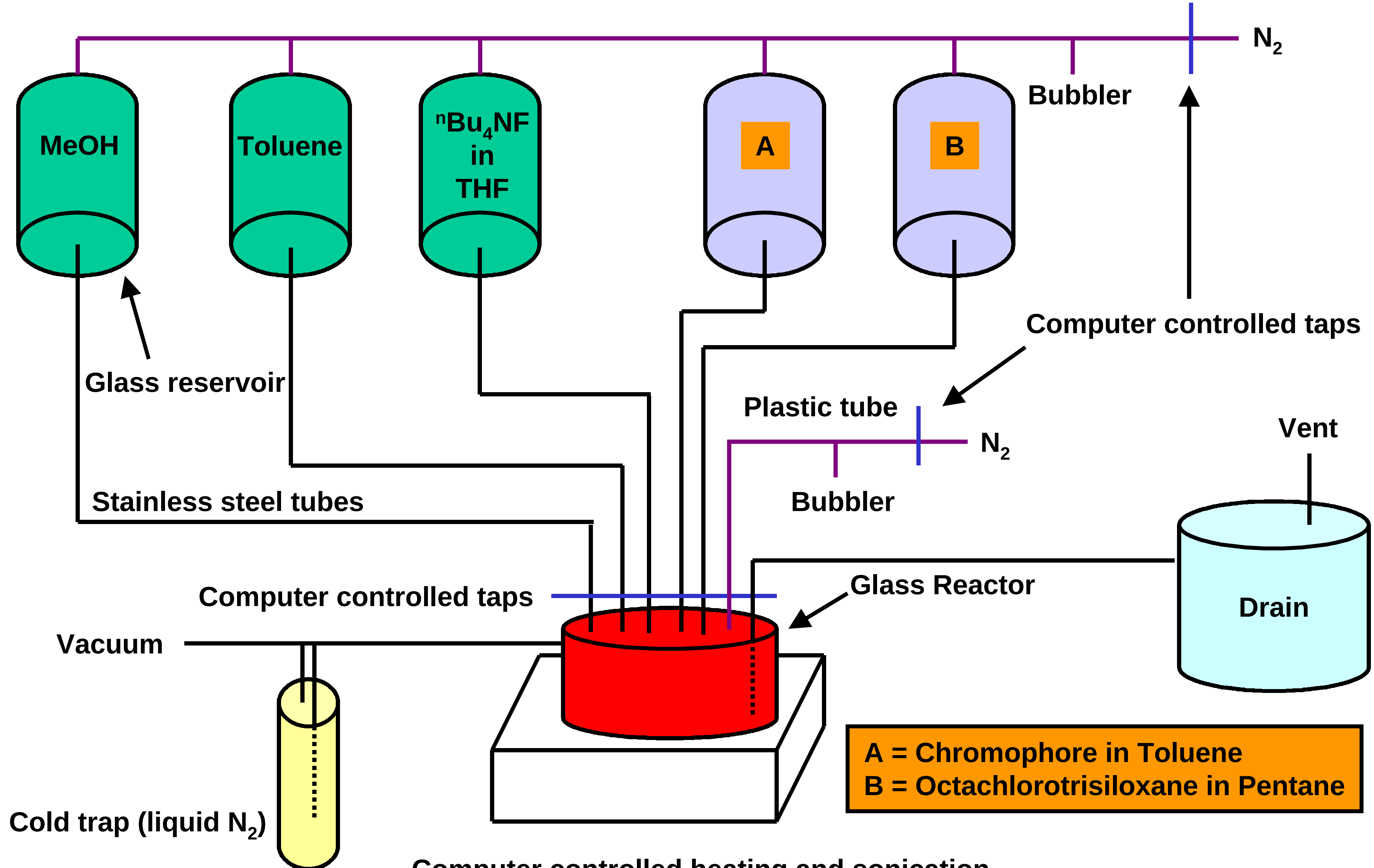
Gallium Oxide formation in 30 min at room temp. from commercially available precursor



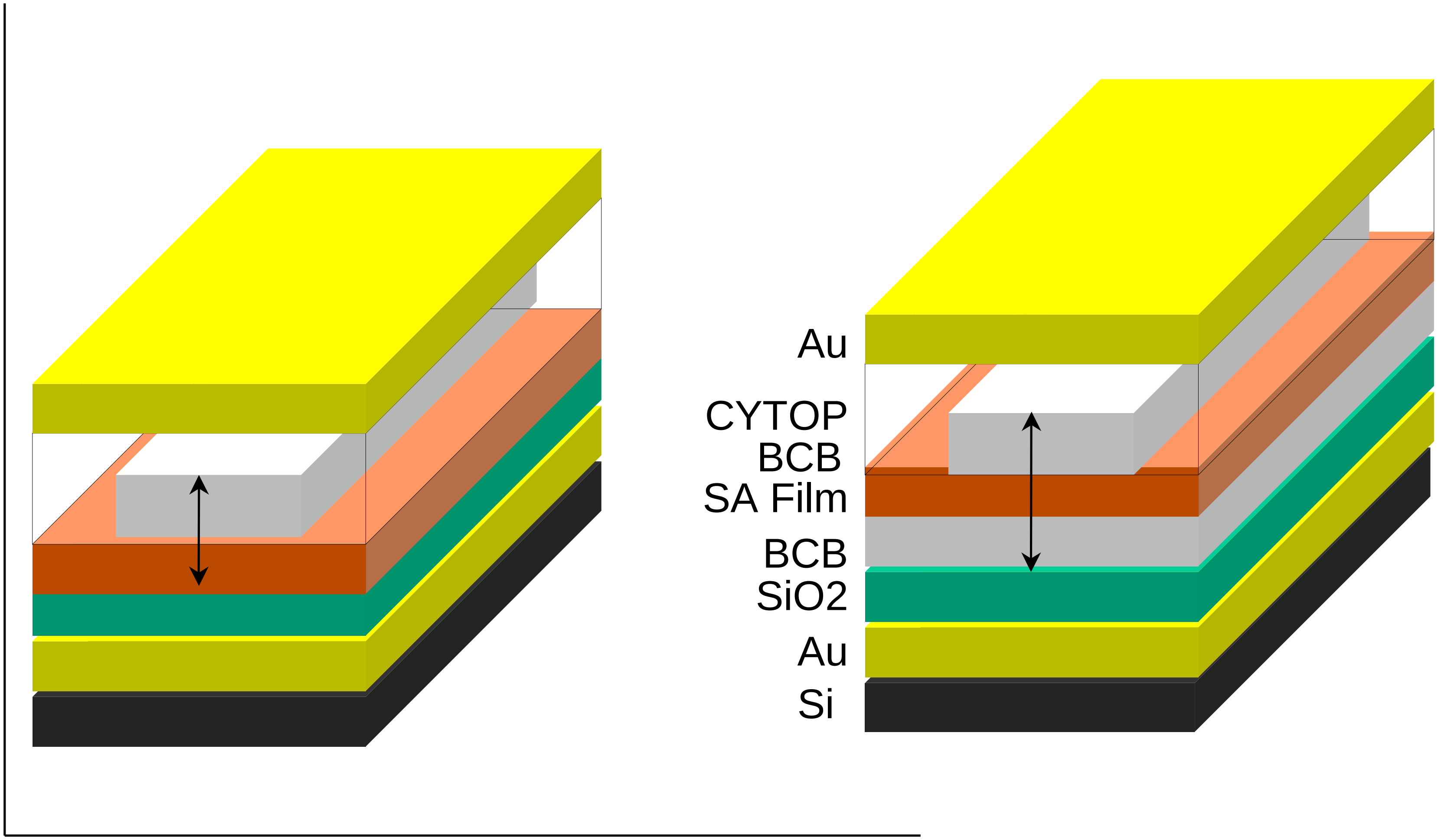


Second Generation Self-Assembly (Protection-Deprotection)

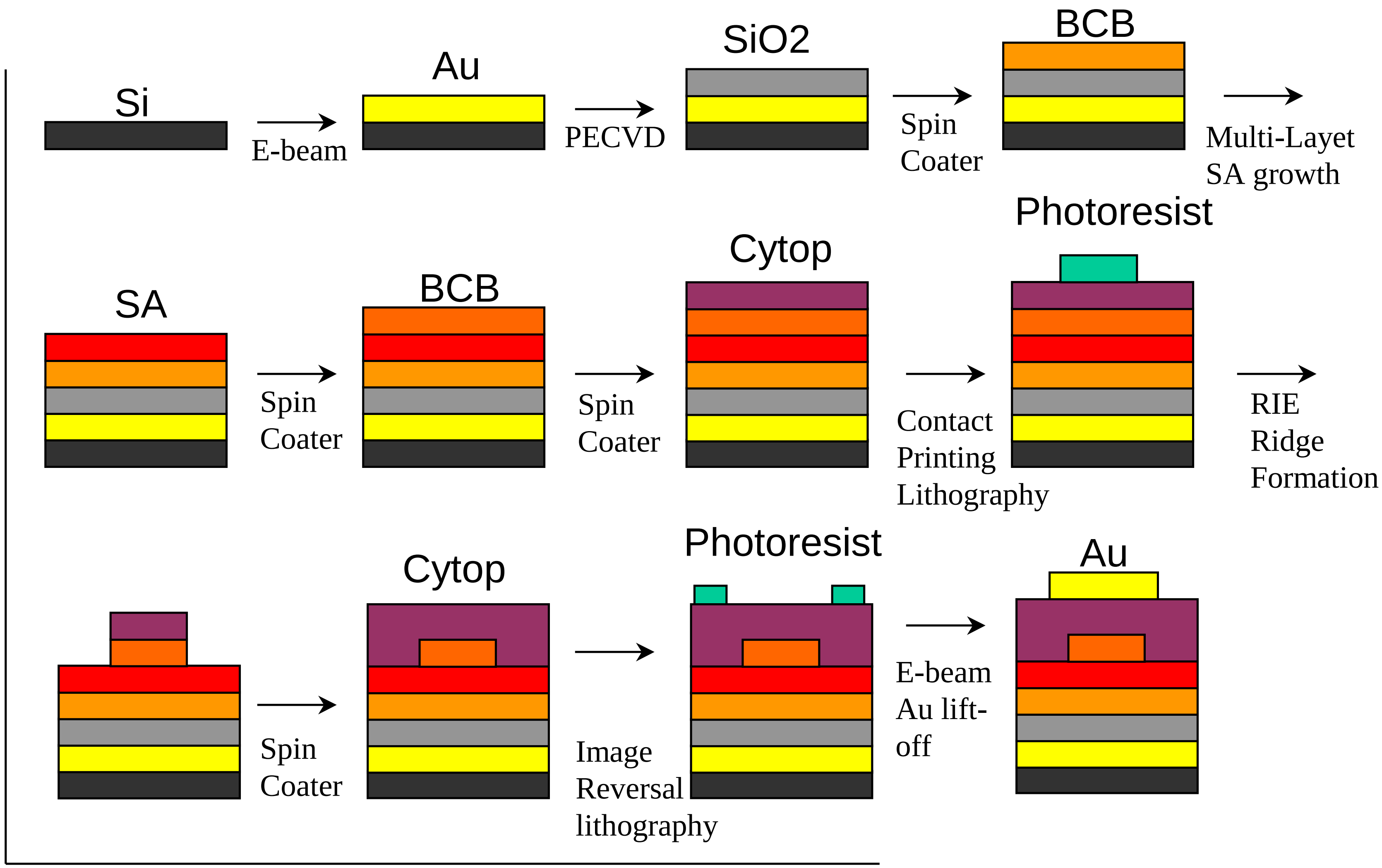
One "Pot"-Chemistry



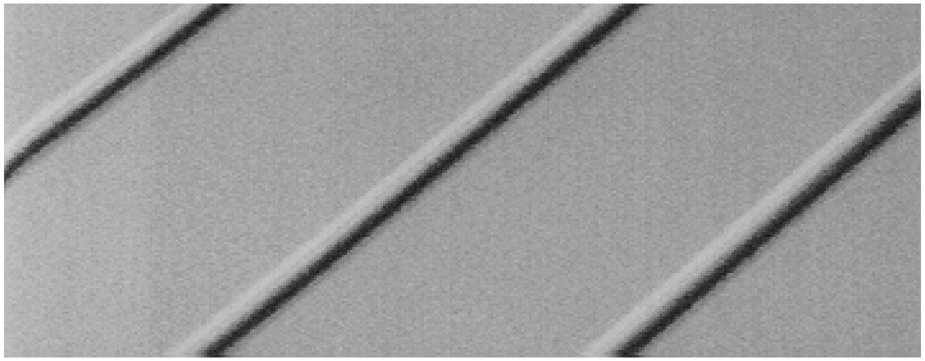
Prototype Channel Waveguide E-O Modulators Using Self-Assembled Organic Superlattices

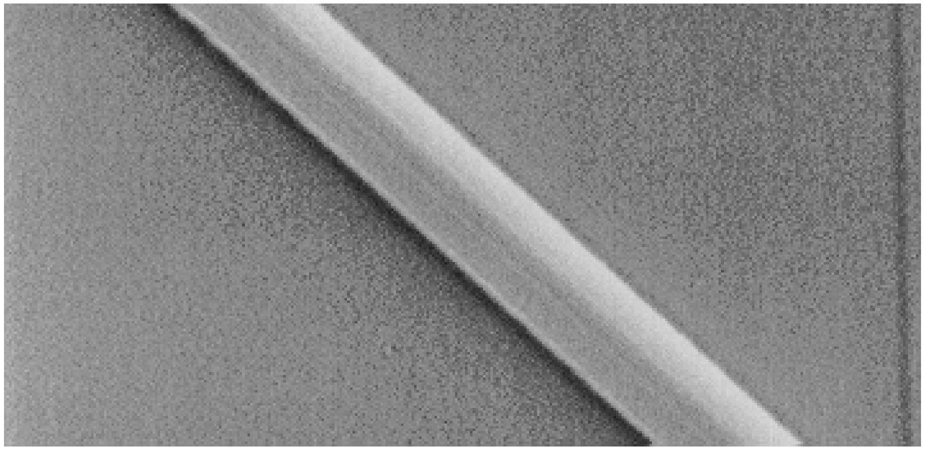


Fabrication Processes of Prototype SA Channel Waveguide E-O Modulators (BCB Sandwich)



SEM of Waveguide Sidewall Roughness

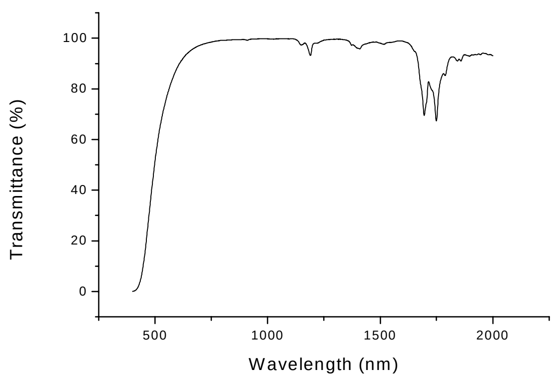




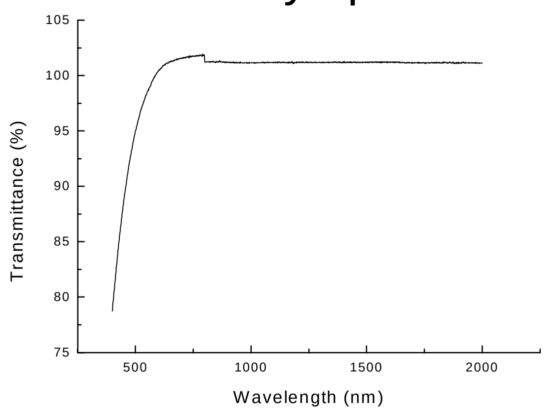
UV-VIS-NIR Transmission Spectra

μm\ α (cm-1)	BCB	Cytop
1.064	0.044	0.035
1.3	0.083	0.039
1.55	0.227	0.023

BCB

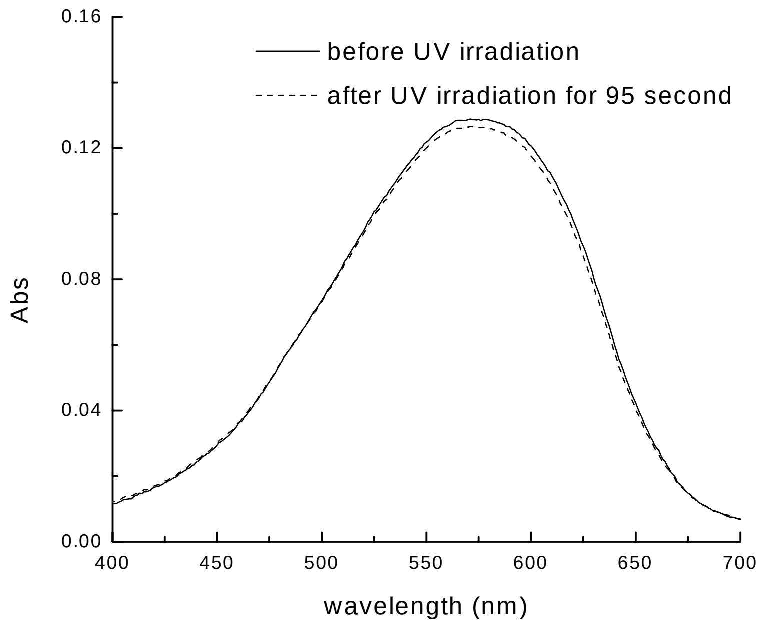


Cytop



Possible Impact During Modulator Processing -- Photolithography

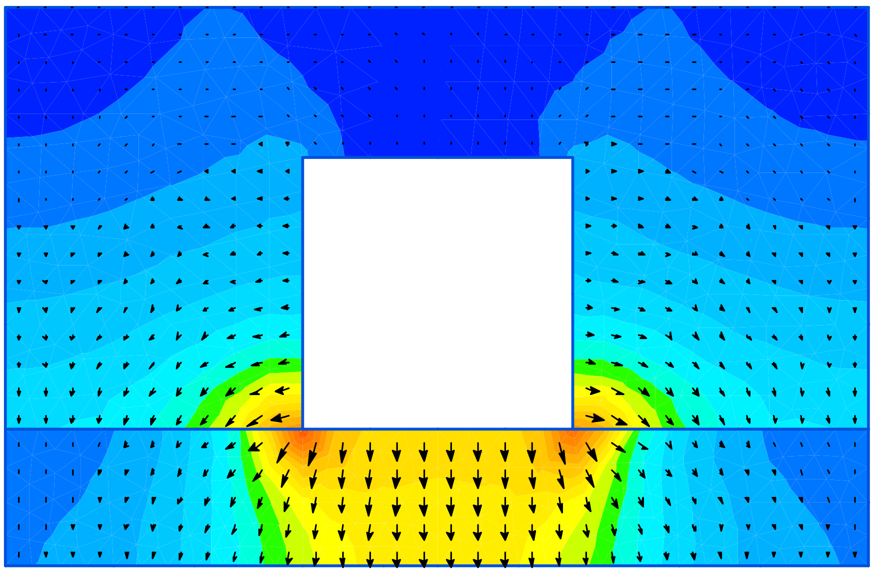
UV-visible absorption



SHG

$I^{1/2}$: 1.410 (before UV irradiation)
$I^{1/2}$: 1.401 (after UV irradiation)

1.2 μm self-assembly with 2μm buffer layer



Width = 10 μm

Thickness = 10 μm

$V_\pi L \approx$ 1.56V.cm

f_{BW}(walk-off).L
= 50GHz.cm

Z = 50 ohms

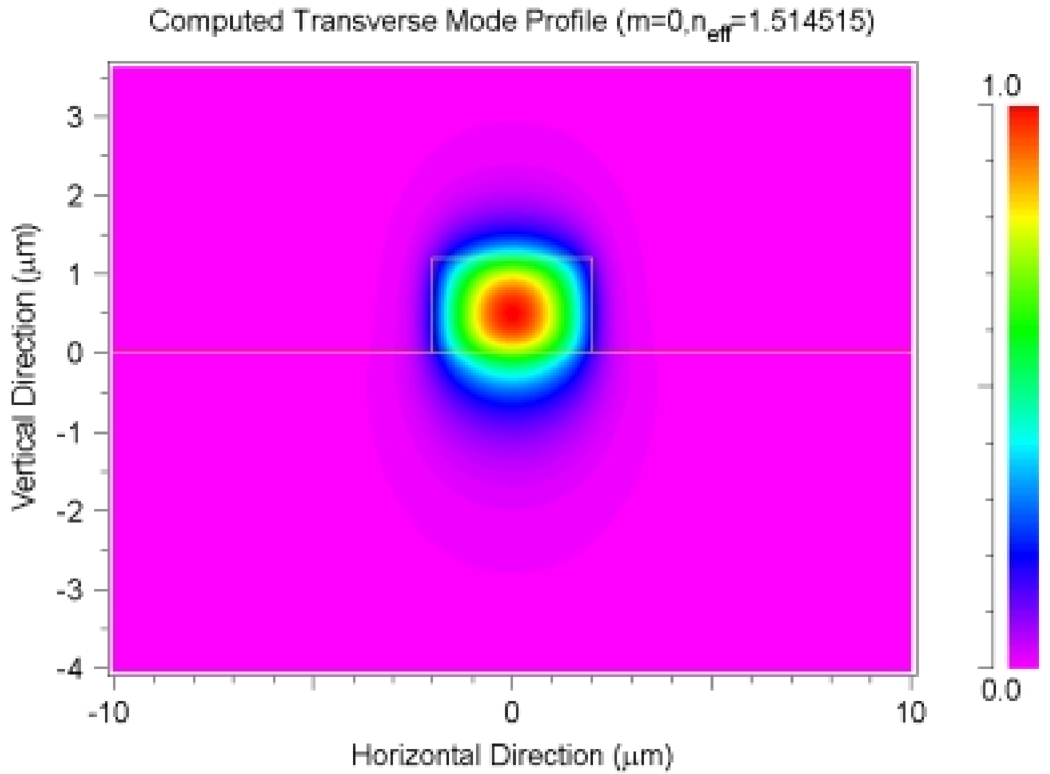
R33=56pm/V

Operating (λ=1.3-1.6 μm)

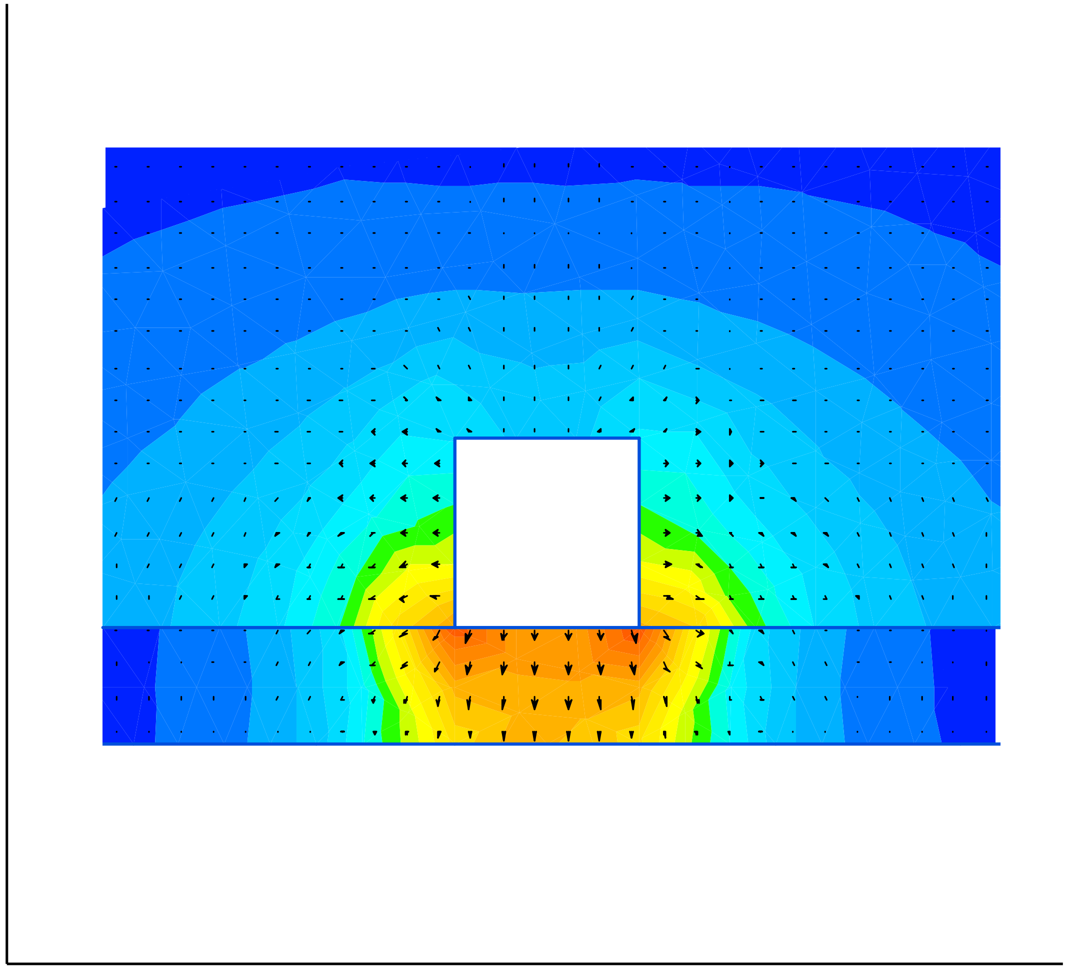
50GHz V_π = 1.56V

100GHz V_π = 3.12V

200GHz V_π = 6.24V


Computed Transverse Mode Profile (m=0,n_eff=1.514515)
Vertical Direction (µm)
3
2
1
0
-1
-2
-3
-4
-10
0
10
Horizontal Direction (µm)
1.0
0.0

0.6 μm self-assembly with 1μm buffer layer



Width = 4 μm

Thickness = 4 μm

$V_\pi L \approx 0.78$V.cm

f_{BW}(walk-off).L

= 56GHz.cm

Z = 50 ohms

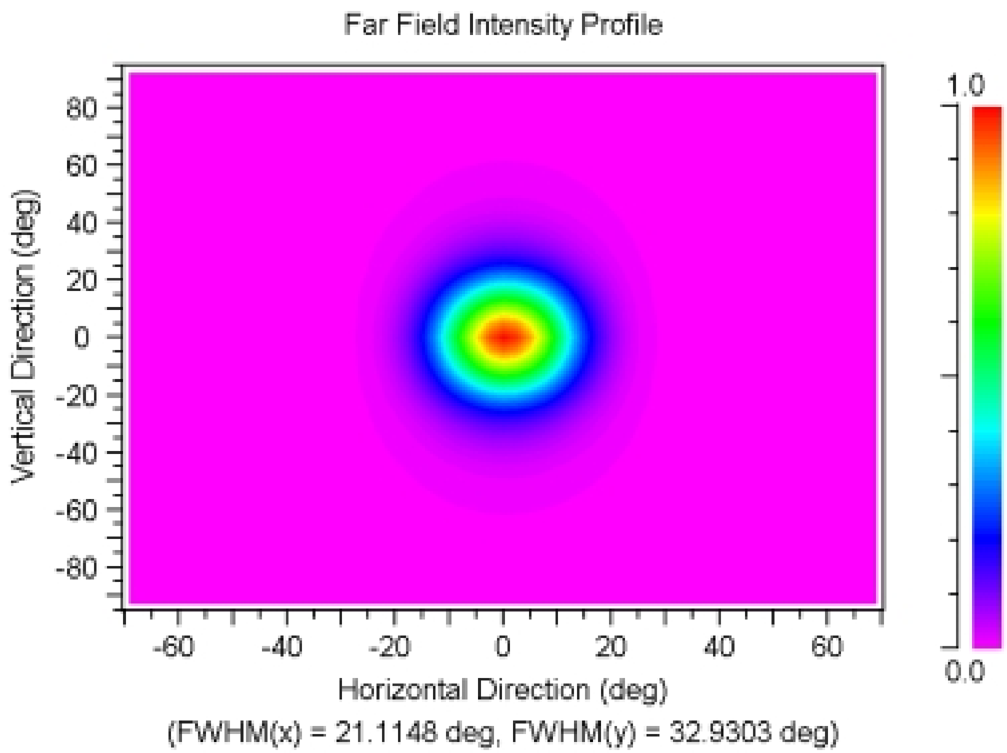
R33=56pm/V

Operating (λ=1.3-1.6 μm)

32GHz V_π = 0.78V

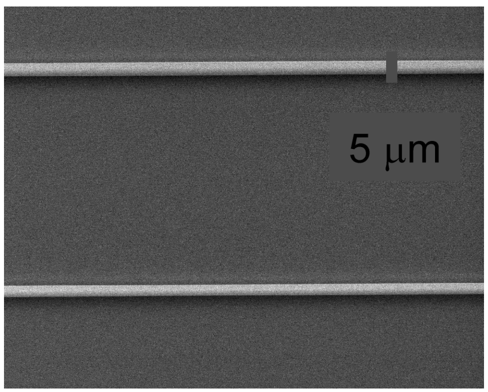
112GHz V_π= 1.56V

224GHz V_π= 3.12V

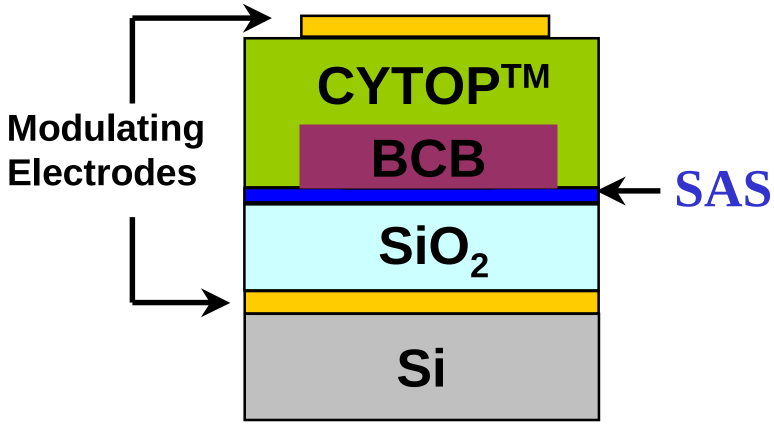

Far Field Intensity Profile
Vertical Direction (deg)
80
60
40
20
0
-20
-40
-60
-80
-60
-40
-20
0
20
40
60
Horizontal Direction (deg)
(FWHM(x) = 21.1148 deg, FWHM(y) = 32.9303 deg)
1.0
0.0

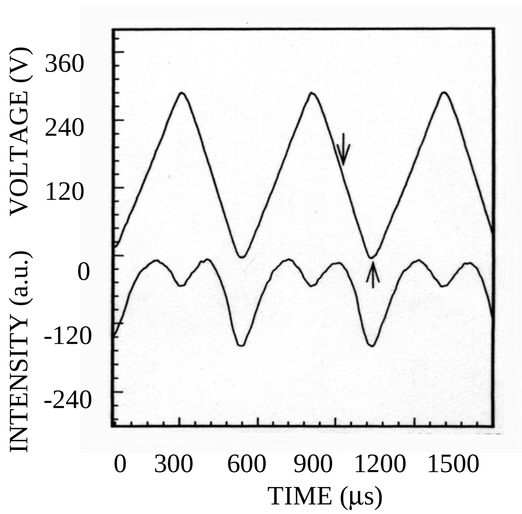
THE FIRST SELF-ASSEMBLED ELECTRO-OPTIC MODULATOR

- Self-Assembly / Growth Directly on Substrate
- No Poling, No Electrically Matched Buffer Layer
- Cladding Layers Commercially Available, Electronic Grade Polymers
- Stable at 80 ^{o}C



SEM





For 40 Layer Device, First Generation Chromophore, r_{33}^{eff} = 56 pm/V

RESEARCH AGENDA

- Second-Generation Chromophore, Second-Generation Assembly
- BCB Above and Below SAS, Measure Loss
- Longer, Thicker SAS → $V_{\pi} < 4$ V
- Transparent Conducting Oxide Modulating Electrodes

Future Efforts

SA Organic EO modulators

1. Materials
   - Implement Automated Assembly
   - Implement New Super Chromophores
   - Characterize EO, Loss Characteristics

2. Device
   - Waveguide Fabrication, Testing
   - Modulator Fabrication, Testing